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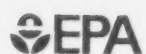
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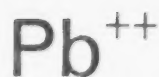
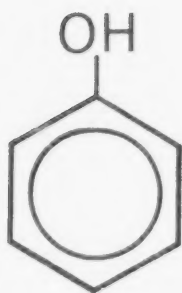
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Health Effects of Land Treatment: Toxicological

by

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FOREWORD

The many benefits of our modern, developing, industrial society are accompanied by certain hazards. Careful assessment of the relative risk of existing and new man-made environmental hazards is necessary for the establishment of sound regulatory policy. These regulations serve to enhance the quality of our environment in order to promote the public health and welfare and the productive capacity of our Nation's population.

The complexities of environmental problems originate in the deep interdependent relationships between the various physical and biological segments of man's natural and social world. Solutions to these environmental problems require an integrated program of research and development using input from a number of disciplines. The Health Effects Research Laboratory, Research Triangle Park, NC and Cincinnati, OH, conducts a coordinated environmental health research program in toxicology, epidemiology, and clinical studies using human volunteer subjects. Wide ranges of pollutants known or suspected to cause health problems are studied. The research focuses on air pollutants, water pollutants, toxic substances, hazardous wastes, pesticides, and nonionizing radiation. The laboratory participates in the development and revision of air and water quality criteria and health assessment documents on pollutants for which regulatory actions are being considered. Direct support to the regulatory function of the Agency is provided in the form of expert testimony and preparation of affidavits as well as expert advice to the Administrator to assure the adequacy of environmental regulatory decisions involving the protection of the health and welfare of all U.S. inhabitants.

This report provides a general appraisal of the impact of toxicological contaminants in wastewater when applied to land. It is assumed that only a minimum of preapplication treatment is given so that the land itself serves as part of the treatment system. With a better understanding of such factors as the toxic substances present in wastewater, decomposition rates, and toxicology, more informed decisions may be made on proper management practices necessary to protect public health in the community.

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Director
Health Effects Research Laboratory

ABSTRACT

The potential health effects arising from the land treatment of wastewater are examined, and an appraisal of these effects made. The agents, or pollutants, of concern from a health effects viewpoint are divided into the categories of pathogens and toxic substances. Only the latter are considered in this volume, the former having been discussed in a previous volume. The toxic substances include organics, trace elements, nitrates, and sodium. These agents form the basis of the main sections of this report.

For each agent of concern the types and levels commonly found in municipal wastewater and the efficiency of preapplication treatment (usually stabilization pond) are briefly reviewed. A discussion of the levels, behavior, and survival of the agent in the medium or route of potential human exposure, i.e., aerosols, surface soil and plants, subsurface soil and groundwater, and animals, follows as appropriate. Finally, conclusions and research needs are presented.

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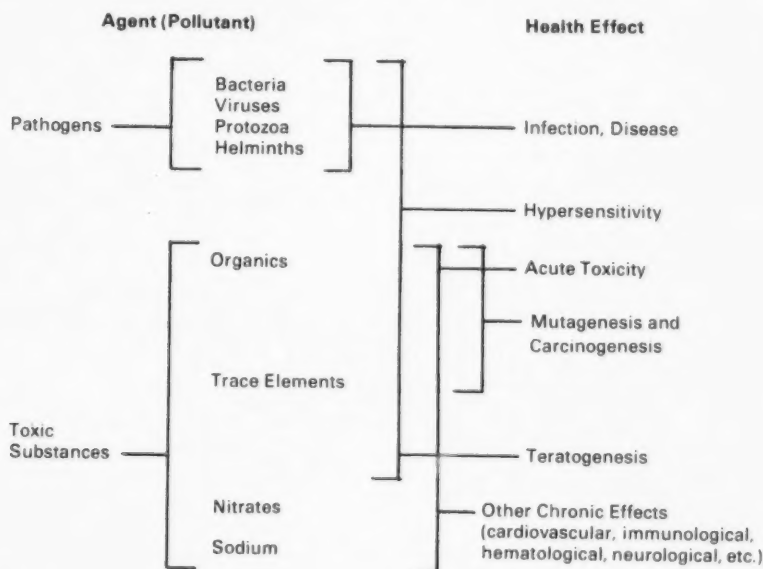
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INTRODUCTION

It is the purpose of this report to examine the potential health effects of land treatment, and to provide an appraisal of these effects. The agents, or pollutants, of concern from a health effects viewpoint can be divided into the two broad categories of pathogens and toxic substances. The pathogens have been covered in an earlier report, "Health Effects of Land Treatment: Microbiological" (Kowal 1982). That report also contains more general introductory material on land treatment, and is to be considered a companion report to this one. The pathogens include bacteria (e.g., *Salmonella* and *Shigella*), viruses (i.e., enteroviruses, hepatitis virus, adenoviruses, rotaviruses, and Norwalk-like agents), protozoa (e.g., *Entamoeba* and *Giardia*), and helminths (or worms, e.g., *Ascaris*, *Trichuris*, and *Toxocara*). The protozoa and helminths are often grouped together under the term, "parasites," although in reality all the pathogens are parasites. The toxic substances include organics, trace elements (or heavy metals, e.g., cadmium and lead), nitrates, and sodium. Nitrates and sodium are usually not viewed as "toxic" substances, but are here so considered because of their potential hematological and long-term cardiovascular effects when present in water supplies at high levels. These agents form the basis of the main sections of this report. The major health effects of these agents are listed below:



For each agent of concern the types and levels commonly found in municipal wastewater and the efficiency of preapplication treatment (usually stabilization pond) are briefly reviewed. A discussion of the levels, behavior, and survival of the agent in the medium or route of potential human exposure, i.e., aerosols, surface soil

and plants, subsurface soil and groundwater, and animals, follows as appropriate. Finally, conclusions and research needs are presented.

Surface water pollution from land treatment site runoff is not considered since proper system design should prevent direct runoff to surface waters (Sorber and Guter 1975, Reed 1979, USEPA 1981). Surface discharge of overland flow effluent may have similar consequences to those of conventional treatment, but little is known in this area since examples are so few.

The present volume is devoted to the toxic substances. For a discussion of the pathogens see Kowal (1982).

ORGANICS

The potential health effects of toxic organic compounds are myriad. Systems affected range from the dermatological to the nervous to the subcellular, and effects produced range from rash to motor dysfunction to cancer. The degree of toxicity of organic compounds varies widely, from essentially harmless (e.g., most carbohydrates) to moderately toxic (e.g., most alcohols) to extremely toxic (e.g., aflatoxins).

A glance at the current edition of The Merck Index will reveal that the number of organic compounds described thus far is almost unfathomable. Nearly any of these may appear in wastewater, depending upon its sources. Thus, the discussion below must be perforce rather general, and the presence of any particular toxic organic in high concentration in the wastewater may require a site-specific evaluation of potential health effects.

Types and Levels in Wastewater

Most common organics in domestic wastewater derive from feces, urine, paper products, food wastes, detergents, and skin excretions and contaminants (from bathing). In medium-strength sewage (700 ppm solids content), organics make up about 75% of the suspended solids and about 40% of the filterable solids (colloidal and dissolved), consisting primarily of proteins (40-60%), carbohydrates (25-50%), and fats and oils (10%) (Metcalf, and Eddy 1972). After secondary treatment, the more refractory and high molecular-weight organics predominate, e.g., fulvic acid, humic acid, and humathomelanic acid (Chang and Page 1978). In general, however, the chemical nature of domestic wastewater remains poorly characterized.

Although most of the organics found in domestic wastewater are probably harmless in a land treatment context, it has recently been found that fecal material commonly contains mutagens. Thus, there is evidence that one of the causes of colorectal cancer is the presence of carcinogens or co-carcinogens produced by the bacterial degradation in the gut of bile acids or cholesterol (Thornton 1981). The mutagenicity of feces can be increased by anaerobic incubation and by the presence of bile and bile acids (Van Tassell *et al.* 1982), and the level of mutagenicity generally is lower in vegetarians than non-vegetarians (Kuhnlein *et al.* 1981). High levels of chromosome-breaking mutagenic activity have also been found in the feces of animals—dog, otter, gull, cow, horse, sheep, chicken, and goose (Stich *et al.* 1980). The chemical nature of the fecal mutagens is unknown. In the case of the latter animal mutagens, evidence suggests that at least part of the mutagenic action is due to hydrogen peroxide and the ensuing radicals which can be formed during oxidation of many organic compounds (Stich *et al.* 1980).

In the State of Illinois ten domestic and industrial secondary effluents were examined for mutagenicity by Johnston *et al.* (1982), with the results that all ten effluents assayed showed significant mutagenicity. Mutagenic activity per unit volume of effluent varied over a 4,500-fold range, and toxicity varied over a 120-fold range. Selective extraction of whole effluents appeared to unmask mutagenic activity, probably by separating mutagens and substances that interfere with the mutagen assay. In several effluents there was evidence of several mutagenic compounds present, and it appeared that the mutagens were predominantly nonpolar, neutral compounds. There was no obvious influence of disinfection by chlorination on the effluent mutagenicity, in spite of the fact that one would expect many mutagens to be

formed by the action of chlorine on humic substances and other organics found in wastewater.

The major contributors of toxic organics to municipal wastewaters are usually assumed to be industrial discharges. However, household wastewater discharge may represent an important contributor since many consumer products in daily use contain toxic substances. A recent study (Hathaway 1980) identified consumer products containing toxic compounds on EPA's list of 129 priority pollutants, which may eventually end up in wastewater. The most frequently used products are cleaning agents and cosmetics, containing solvents and heavy metals as main ingredients. Next are deodorizers and disinfectants, containing naphthalene, phenol, and chlorophenols. Discarded into wastewater infrequently, but in large volumes, are pesticides, laundry products, paint products, polishes, and preservatives. The organic priority pollutants most frequently used and discharged into domestic wastewater were predicted to be the following:

benzene	naphthalene
phenol	toluene
2,4,6-trichlorophenol	diethylphthalate
2-chlorophenol	dimethylphthalate
1,2-dichlorobenzene	trichloroethylene
1,4-dichlorobenzene	aldrin
1,1,1-trichloroethane	dieldrin

Because of the difficulty of analysis of complex mixtures, it has only recently been possible to measure the actual levels of organics in wastewater using advanced methods of extraction, gas and other chromatography, mass spectrometry, and computer analysis. The U. S. Environmental Protection Agency has sponsored two extensive surveys of the types and levels of priority pollutants in municipal wastewaters, which, of course, result from both domestic and industrial discharges. The first (DeWalle *et al.* 1981), supported by the Municipal Environmental Research Laboratory in Cincinnati, covered 25 cities located throughout the United States, and the second (Feiler 1980), supported by the Effluent Guidelines Division in Washington, D.C., covered 40 cities, the results from 20 of which are reported in the cited reference.

In the 25-city survey (DeWalle *et al.* 1981) most of the 24-hour composite samples of raw wastewaters contained a total of less than 1 mg/l of priority organics, and the numbers of compounds detected clustered between 20 and 50. In the 40-city survey (Burns and Roe 1982) six days of 24-hour sampling was completed, and the samples from 20 cities were analyzed. The priority organics detected in at least 50% of the samples analyzed in either survey are listed, together with their concentrations, in Table I.

Comparison of the results of the two surveys with the list of organic priority pollutants most likely to be discharged into domestic wastewater, reveals considerable overlap, and gives one some confidence that these two studies have yielded a reasonable characterization of the priority organics in municipal wastewater, at least of those identifiable by modern methods. The broad range of concentrations detected among the samples, however, suggests that wastewater applied to land should be regularly monitored for toxic organics. This measure is emphasized by the occasional discharge of toxic substances into municipal wastewater systems with resulting medical effects in treatment plant workers, such as the recent hexachlorocyclopentadiene episode in Louisville, Kentucky (Kaminsky *et al.* 1980).

Preapplication Treatment

Temporary storage ponds are usually used with land treatment systems because of the need to (1) remove grit, organic solids, and grease so as to prevent fouling of

Table 1. Most Frequently Detected Priority Organics in Raw Municipal Wastewater

Compound	DeWalle et al. 1981		Burns and Roe 1982	
	Detection Frequency (%)	Concentration Range ($\mu\text{g/l}$)	Detection Frequency (%)	Concentration Range ($\mu\text{g/l}$)
Phenol	94	0.90-2440.00	79	1-1,400
1,1,1-Trichloroethane	94	0.40- 97.50	85	1-30,000
Trichloroethylene	94	0.90-1553.00	90	1-1,800
Tetrachloroethylene	94	1.50- 385.10	95	1-5,700
Ethylbenzene	94	0.20- 304.40	80	1-730
Trichloromethane (Chloroform)	94	0.25- 73.10	91	1-430
Diethylphthalate	91	1.34- 290.00	53	1-42
Di-n-butylphthalate	91	0.26- 123.00	64	1-140
Toluene	90	0.70- 795.00	96	2-1,300
Dichloromethane	90	0.50- 666.10	92	1-49,000
Bis(2-ethylhexyl)phthalate	89	0.06- 117.00	92	2-670
Naphthalene	86	1.25- 291.00	49	1-150
1,4-Dichlorobenzene	83	1.70- 119.00	17	2-200
Phenanthrene	83	0.20- 49.50	20	1-93
Benzene	79	0.26- 243.00	61	1-1,560
Heptachlor	77	0.30- 37.00	5	0.08-0.50
Butylbenzylphthalate	77	1.10- 237.00	57	2-560
BHC-G (Lindane)	71	0.05- 11.20	26	0.02-3.9
1,2-Dichlorobenzene	69	0.78- 703.00	23	1-440
Dimethylphthalate	66	0.09- 114.00	11	1-110

Table 1. (Continued)

Compound	DeWalle et al. 1981		Burns and Roe 1982	
	Detection Frequency (%)	Concentration Range (µg/l)	Detection Frequency (%)	Concentration Range (µg/l)
BHC-D	63	0.01- 5.10	3	0.10-1.4
Dieldrin	63	0.02- 4.40	1	0.03-0.04
1,3-Dichlorobenzene	60	0.08- 548.00	7	2-270
BHC-A	60	0.01- 2.90	8	0.02-4.4
DDT	60	0.10- 24.00	<1	1.2
Di-n-octylphthalate	57	0.31- 15.50	7	2-210
1,1-Dichloroethane	55	0.20- 3.60	31	1-24
1,2-Dichloroethane	55	0.20-3950.00	15	1-76,000
DDD	54	0.05- 10.00	1	0.31-0.77
Anthracene	51	0.04- 36.80	18	1-93
Aldrin	51	0.02- 2.00	1	0.03-5
Endosulfan-B	51	0.20- 8.80	—	—
1,2-Trans-dichloroethylene	20	0.20- 45.30	62	1-200

wastewater distribution system components, (2) provide storage during winter when crop uptake for nitrogen control is minimal and the soil biota may be inaccessible and/or relatively inactive, (3) provide storage during periods of precipitation, and (4) equalize wastewater flows. Storage ponds also serve a treatment function, as was illustrated in the earlier report, on microbiology.

One of the few land treatment sites where the fate of organics has been followed from input to output is that at Muskegon, Michigan (Demirjian *et al.* 1983). Here the treatment train for about 30 MGD of combined industrial (over 70%, including several organic chemical manufacturers) and domestic wastewater consists of 8 acres of aerated lagoons, 850 acres of storage lagoons, and 5,500 acres of sprayirrigated farmland, which is drained by subsurface (5-12 feet) tiles. The detention time of the raw wastewater in the aerated lagoons is approximately 36 hours, and that in the storage lagoons is variable, depending upon the season of the year.

The influent to the Muskegon system generally contains 20-30 priority organic pollutants and 15-25 additional organic compounds. The aerated lagoons remove over 90% of most compounds by stripping of the volatiles, over 99% of toluene and acetone for example. Data have shown variable removal rates for organics in the storage lagoons, sometimes quite poor, but organics average below 10 $\mu\text{g/l}$ for most compounds before spray irrigation. Removals for most compounds are better during summer months than winter.

Aerosols

While most of the nonvolatile organics in wastewater at a land treatment site would be expected to enter the soil, much of the volatile organics would probably be released to the air. The fraction entering the air would depend, of course, upon the processes, detention times, and application rates used at a particular facility. Although some of the volatile organics (and nonvolatile organics) may be released in the form of true aerosols, most would be expected to be released as gases by evaporation (volatilization). The process of air stripping has been shown to be particularly efficient in releasing volatile organics at aeration basins of activated sludge wastewater treatment plants, e.g., the compounds hexachlorobicycloheptadiene, heptachlorobicycloheptene, and chlordane at Memphis, Tennessee (Clark *et al.* 1981). Continuous chronic exposures with intermittent acute exposures to these toxic organics in Memphis may have resulted in significant health effects in the treatment plant workers.

A study at an overland flow site has suggested that volatilization reduces the levels of toxic volatile organics in wastewater by 80-100%, depending on the specific substance and application rate (Jenkins *et al.* 1981). The removal was adequately described by first-order kinetics. The results indicated that removal by sorption on suspended matter and sedimentation was not significant.

A certain amount of evaporation occurs during the act of spraying itself. Thus, at a prototype slow rate land treatment system Jenkins and Palazzo (1981) found a 46% to 69% removal of eight volatile organics during sprinkler application.

At the Muskegon slow-rate land treatment system, the behavior of four of the most common toxic organics was studied (Clark *et al.* 1981): trichloroethane, trichloroethylene, tetrachloroethylene, and chloroform (trichloromethane). Air stripping in the aerated lagoons was shown to be significant for these compounds. The maximum concentrations in air, together with the associated wastewater concentrations, of these compounds immediately downwind of the aerated lagoons and spray irrigation rigs are shown in Table 2. All of these air concentrations are well below the 8-hour occupational standards of 45,000 $\mu\text{g/m}^3$, 535,000 $\mu\text{g/m}^3$, 670,000 $\mu\text{g/m}^3$, and 50,000 $\mu\text{g/m}^3$, respectively (ACGIH 1979).

Comparison of the wastewater concentrations in Table 2 with the maximum values found in Table 1 suggests that an increase of three orders of magnitude over the

Table 2. Air and Wastewater Concentrations at Muskegon Land Treatment System

	Aerated Lagoons		Spray Irrigation Rigs	
	Wastewater ($\mu\text{g/l}$)	Air ($\mu\text{g/m}^3$)	Wastewater ($\mu\text{g/l}$)	Air ($\mu\text{g/m}^3$)
Trichloroethane	ND*	90	3	2.7
Trichloroethylene	118	73	68	9.3
Tetrachloroethylene	8.9	46	40	8.6
Chloroform	480	202	—	ND

*ND = Not Detected ($<0.1 \mu\text{g/l}$).

Muskegon values is the maximum that would ever be likely to occur. Even such an increase would probably still result in acceptable air concentrations.

Soil and Groundwater

Organic compounds in wastewater may be volatilized, immobilized by adsorption, or transported through the soil column, possibly to reach the groundwater. Adsorbed organics may be subsequently chemically or photochemically degraded, microbially decomposed, or desorbed. A considerable body of research has been performed on the behavior of pesticides in soil. This research has shown that the affinity of soil materials for pesticides, and presumably for organics in general, decreases in the following order (Chang and Page 1978):

Organic Matter
Vermiculite
Montmorillonite
Illite
Chlorite
Kaolinite

Iron and aluminum oxides also adsorb organics. Adsorption of organic pesticides tends to increase with the concentration of functional groups such as amine, amide, carboxyl, and phenol. Both laboratory and field experiments suggest that, because of adsorption by soil particles, most pesticide residues remain in surface soils during land treatment (Chang and Page 1978).

It has recently been shown that for polynuclear aromatic hydrocarbons adsorption increases with increasing organic carbon content of the soils and increasing "effective chain length" of the molecule (Means *et al.* 1980). The behavior of polychlorinated biphenyls (PCBs) in soil has been comprehensively reviewed by Griffin and Chian (1980), who concluded that PCBs are strongly adsorbed by soil, and that the nature of the surface, the soil organic matter content, and the chlorine content and/or hydrophobicity of the individual PCB isomers are factors affecting adsorption. Adsorption increases with increasing organic matter content of the soil, with increasing chlorine content, and with increasing hydrophobicity. One study of PCB percolation through soil columns showed that less than 0.05% of one isomer was leached in the worst case.

Once organics are immobilized by adsorption on the surfaces of soil particles, microbial decomposition, or biodegradation, is probably the major mechanism of their breakdown. Although there are several abiotic mechanisms for chemical change, nonenzymatic reactions rarely result in appreciable changes in chemical structure, and it is biodegradation that brings about major alterations and mineralization of organics (Alexander 1981). The chief agents of this metabolism are the indigenous heterotrophic bacteria and fungi.

The potential of microbial decomposition for removal of organics is demonstrated by the experience at two rapid infiltration and one overland flow land treatment sites.

At Flushing Meadows in Arizona secondary effluent has resulted in no accumulation of organic carbon in the soil after ten years of operation and 754 m of total infiltration (Bouwer and Rice 1978). Secondly, the Lake George Village Sewage Treatment Plant in New York has been applying unchlorinated secondary effluent to natural delta sand beds by rapid infiltration since 1939 (Aulenbach and Clesceri 1978). After about forty years of daily infiltration rates of 0.08 to 0.30 m/day, there were no indications that the soil's capacity to treat the applied effluent was approaching exhaustion. The greatest removal of constituents occurred in the top 10 m of the sand beds. At a prototype overland flow land treatment system, at the U.S. Army Cold Regions Research and Engineering Laboratory in New Hampshire, greater than 94% removal of each of 13 trace organics by volatilization and adsorption was observed (Jenkins *et al.* 1983), with removal efficiencies decreasing as application rates increased and temperature decreased. With the possible exception of PCB, biodegradation resulted in the absence of contaminant buildup in the surface soil.

Although complete mineralization and detoxication of organic compounds is common, many compounds are acted on by microorganisms in soils even though these microorganisms are unable to use them as their sources of nutrient or energy. The microorganisms are probably utilizing another substrate while performing the transformations known as "cometabolism" (Alexander 1981). Cometabolism may lead to detoxication, the formation of new toxic substances, or the synthesis of persistent products. There is evidence that cometabolism may be particularly common for toxic organics in very low concentrations in the environment (Rubin *et al.* 1982, Subba-Rao *et al.* 1982).

The metabolism of few chemicals has been studied in microbial cultures, and even fewer in natural ecosystems. Why certain intermediate compounds in a metabolic sequence accumulate outside or inside the active organism is not known, and it is extremely difficult to predict the chemical fate of toxic organics in the environment. The prediction of biodegradability from chemical structure, although theoretically possible, has thus far proven problematic. Alexander (1981) has described several common types of reactions that may occur, and these are listed in Table 3. It used to be thought that every organic compound could be completely decomposed by microorganisms. Thus a recent evaluation (Kobayashi and Rittmann 1982) indicated that the use of properly selected populations of microorganisms, and the maintenance of appropriate controlled environmental conditions, could be an important means of improving biological treatment of organic wastes, and that members of almost every class of synthetic compound can be degraded by some

Table 3. Common Types of Chemical Transformations in the Environment

Dehalogenation	Nitro metabolism
Deamination	Oxime metabolism
Decarboxylation	Nitrile/amide metabolism
Methyl oxidation	Cleavage reactions (many types)
Hydroxylation and ketone formation	Nondegradative reactions:
β oxidation	Methylation
Epoxide formation	Ether formation
Nitrogen oxidation	N-Acylation
Sulfur oxidation	Nitration
=S to =O	N-Nitrosation
Sulfoxide reduction	Dimerization
Reduction of triple bond	Nitrogen heterocycle formation
Reduction of double bond	Oligomer and polymer formation
Hydration of double bond	

microorganism. However, field evidence has resulted in the conclusion that some synthetic organics are decomposed slowly, if at all, and may persist for long periods in the environment. Alexander (1981) has summarized the possible reasons for this.

A general idea of the relative degree of biodegradation of toxic organics to be expected in soil may be gained from Tabak *et al.*'s (1981) studies on organic priority pollutants. They collected data on the biodegradability and rate of microbial acclimation of 96 compounds (5 and 10 mg/l) in a static culture flask screening procedure, using domestic wastewater inoculum and synthetic medium. Microbial acclimation, or adaptation, was measured by making three weekly subcultures; percentage biodegradation was measured after seven days incubation in the dark at 25°C. Their overall results are summarized in Table 4. Significant biodegradation was found for phenolic compounds, phthalate esters, naphthalenes, and nitrogenous organics; variable results were found for monocyclic aromatics, polycyclic aromatics, polychlorinated biphenyls, halogenated ethers, and halogenated aliphatics; and no significant biodegradation was found for organochlorine pesticides.

Extrapolation of the above results to the behavior of toxic organics in the soil must be done with two provisos: (1) biodegradation in soil may be somewhat different from that in the aquatic medium used for the tests, and (2) the lower concentration of the organics at the land treatment site may not elicit microbial activity or enzyme induction. Nevertheless, a comparison of the results with the compounds to be expected in wastewater, as listed in Table 1, is instructive. Among the top ten compounds in the table, nine have significant degradation, and one has slow to moderate degradation with significant volatilization. Among the next ten compounds, eight have significant degradation, two of which are followed by toxicity. Only two compounds are not significantly degraded, the pesticides heptachlor and lindane.

Other than for pesticides, the literature on the microbial decomposition of toxic organics in soil is sparse. The degradation of petroleum hydrocarbons, a mixture of aliphatic, aromatic, and asphaltic compounds, has been reviewed by Atlas (1980, 1981). Factors which appear to be important in encouraging high decomposition rate of petroleum hydrocarbons are high temperature, low concentrations, high soil fertility, and an aerobic environment. There is little evidence for significant downward leaching of oil. Experiments with the high-rate application of high petroleum hydrocarbon sludge to land have shown a 77% degradation rate near the surface after one year, most of the degraded compounds being n-alkanes (Lin 1980), and it was concluded that sludge land disposal would not result in petroleum hydrocarbon buildup in the soil. In studies of organic substances in wastewaters used for irrigation, Dodolina *et al.* (1976) found acetaldehyde, crotonaldehyde, benzaldehyde, cyclohexanone, cyclohexanol, and dichloroethane to disappear from soil within ten days. The biodegradation in soil of polychlorinated biphenyls was reviewed by Griffin and Chian (1980) who concluded that they are degradable, but that resistance increases as isomers have higher chlorination. Polybrominated biphenyls, on the other hand, have shown little biodegradation after one year in soil (Jacobs *et al.* 1978).

In view of the multitudinous variety of organic compounds existing, it is difficult to generalize about their biodegradation in soil. It appears, however, that most organics do become microbially decomposed in the soil, at least to some extent. This is especially true of naturally-occurring compounds, or those resembling them, because of the eons of evolution that have developed microbial enzyme systems to do the job. The more structurally complex the molecule is, e.g., condensed rings or dense branching, and more halogenated it is, the more difficult is biodegradation. A convenient sampling of the international literature on decomposition of organics in soil may be found in Overcash (1981).

It has recently been found that it might be possible for carcinogenic and teratogenic nitrosamines to be formed from secondary and tertiary amines at land treatment sites. Thus, Thomas and Alexander (1981) have shown that dimethylamine

Table 4. Biodegradability of Priority Organic Compounds (after Tabak *et al.* 1981)

D—Significant degradation with rapid adaptation
 A—Significant degradation with gradual adaptation
 T—Significant degradation with gradual adaptation followed by a deadaptive process (toxicity)
 B—Slow to moderate biodegradative activity, concomitant with significant rate of volatilization
 C—Very slow biodegradative activity, with long adaptation period needed
 N—Not significantly degraded under the conditions of test method

Test Compound	Performance Summary	Test Compound	Performance Summary
<i>Phenols</i>			
Phenol		p-Chloro-m-cresol	D
2-Chloro phenol	D	2-Nitro phenol	D
2,4-Dichloro phenol	D	4-Nitro phenol	D
2,4,6-Trichloro phenol	D	2,4-Dinitro phenol	D
Pentachloro phenol	A	4,6 Dinitro-o-cresol	N
2,4 Dimethyl phenol	D		
<i>Phthalate Esters</i>			
Dimethyl phthalate	D	Bis-(2-ethyl hexyl) phthalate	A
Diethyl phthalate	D	Di-n-octyl phthalate	A
Di-n-butyl phthalate	D	Butyl benzyl phthalate	D
<i>Naphthalenes</i>			
Naphthalene	D		
2-Chloro naphthalene	D		
Acenaphthene	D		
Acenaphthylene	D		

Table 4. (Continued)

D—Significant degradation with rapid adaptation
A—Significant degradation with gradual adaptation
T—Significant degradation with gradual adaptation followed by a deadaptive process (toxicity)
B—Slow to moderate biodegradative activity, concomitant with significant rate of volatilization
C—Very slow biodegradative activity, with long adaptation period needed
N—Not significantly degraded under the conditions of test method

Test Compound	Performance Summary	Test Compound	Performance Summary
<i>Monocyclic Aromatics</i>			
Benzene	D	Hexachlorobenzene	N
Chlorobenzene	D-A	Nitrobenzene	D
1,2-Dichlorobenzene	T	Ethylbenzene	D-A
1,3-Dichlorobenzene	T	Toluene	D
1,4-Dichlorobenzene	T	2,4-Dinitrotoluene	T
1,2,4-Trichlorobenzene	T	2,6-Dinitrotoluene	T
<i>Polycyclic Aromatics (PAHs)</i>			
Anthracene	A	1,2-Benzanthracene	N
Phenanthrene	D	Pyrene	D-N
Fluorene	A	Chrysene	A-N
Fluoranthene	A-N		
<i>Polychlorinated Biphenyls (PCBs)</i>			
Aroclor-1016	N	Aroclor-1248	N
Aroclor-1221	D	Aroclor-1254	N
Aroclor-1232	D	Aroclor-1260	N
Aroclor-1242	N		

Table 4. (Continued)

D—Significant degradation with rapid adaptation
A—Significant degradation with gradual adaptation
T—Significant degradation with gradual adaptation followed by a deadaptive process (toxicity)
B—Slow to moderate biodegradative activity, concomitant with significant rate of volatilization
C—Very slow biodegradative activity, with long adaptation period needed
N—Not significantly degraded under the conditions of test method

Test Compound	Performance Summary	Test Compound	Performance Summary
<i>Halogenated Ethers</i>			
Bis-(2-chloroethyl) ether	D	4-Bromodiphenyl ether	N
2-Chloroethyl vinyl ether	D	Bis-(2-chloroethoxy) methane	N
4-Chlorodiphenyl ether	N	Bis-(2-chloroisopropyl) ether	D
<i>Nitrogenous Organics</i>			
<i>Nitrosamines</i>			
N-Nitroso-di-N-propylamine	N	Acrylonitrile	D
N-Nitrosodiphenylamine	D-A	Acrolein	D
<i>Substituted benzenes</i>			
Isophorone	D		
1,2-Diphenylhydrazine	T		
<i>Halogenated Aliphatics</i>			
<i>Chloroethanes</i>			
1,1-Dichloroethane	A	Chloroethylenes	
1,2-Dichloroethane	B	1,1-Dichloroethylene	A
1,1,1-Trichloroethane	B	1,2-Dichloroethylene-cis	B
1,1,2-Trichloroethane	C	1,2-Dichloroethylene-trans	B
1,1,2,2-Tetrachloroethane	N	Trichloroethylene	A
		Tetrachloroethylene	A

Table 4. (Continued)

D—Significant degradation with rapid adaptation
A—Significant degradation with gradual adaptation
T—Significant degradation with gradual adaptation followed by a deadaptive process (toxicity)
B—Slow to moderate biodegradative activity, concomitant with significant rate of volatilization
C—Very slow biodegradative activity, with long adaptation period needed
N—Not significantly degraded under the conditions of test method

Test Compound	Performance Summary	Test Compound	Performance Summary
Hexachloroethane		Chloropropanes	
Halomethanes		1,2-Dichloropropane	A
Methylene chloride	D	Chloropropylenes	
Bromochloromethane	D	1,3-Dichloropropylene	A
Carbon tetrachloride	D	Chlorobutadienes	
Chloroform	A	Hexachloro-1,3-butadiene	D
Dichlorobromomethane	A	Chloropentadienes	
Bromoform	A	Hexachlorocyclopentadiene	D
Chlorodibromomethane	N		
Trichlorofluoromethane	N		
<i>Organochlorine Pesticides</i>			
Aldrin	N	Heptachlor	N
Dieldrin	N	Heptachlor epoxide	N
Chlordane	N	Hexachlorocyclohexane	
DDT p,p'	N	α BHC-alpha	N
DDE p,p'	N	Hexachlorocyclohexane	
DDD p,p'	N	β BHC-beta	N
Endosulfan-alpha	N	Hexachlorocyclohexane	
Endosulfan-beta	N	δ BHC-delta	N
Endosulfan sulfate	N	Hexachlorocyclohexane	
Endrin	N	γ BHC-gamma (lindane)	N

and trimethylamine can be formed in municipal wastewater from naturally-occurring precursors. Dimethylamine may then go on to be microbially nitrosated, forming N-nitrosodimethylamine, a process which can occur under conditions resembling land treatment of wastewater (Greene *et al.* 1981). Whether this can actually occur under field conditions, resulting in a threat to groundwater, has not been demonstrated.

The transport to groundwater of organics has been studied at only a few operating land treatment sites. Bouwer and Rice (1978) have noted that the total organic carbon concentration of the renovated water at the Flushing Meadows rapid infiltration site, in Phoenix, Arizona, averaged about 4 mg/l, indicating the presence of refractory trace organics. They felt that these organics were the main health concern for possible potable use of the groundwater, since their identity is not completely known and carcinogenicity or other toxicity is suspected. More detailed data on groundwater contamination by organics is now available from the nearby 23rd Avenue Project rapid infiltration site, also in Phoenix, Arizona (Tomson *et al.* 1981). At this site secondary effluent is treated by about 25 feet of soil. The overall removal rate of refractory volatile organics (RVO) at this site was 86.4%, resulting in a groundwater concentration of RVO of 2.245 $\mu\text{g/l}$. Removal rates are presented, by class, in Table 5. The most likely volatile organic contaminants of groundwater at this site are the chloroalkanes, alkylphenols, alkanes, and amides.

Table 5. Removal Efficiencies of Refractory Volatile Organics at 23rd Avenue Project Rapid Infiltration Site (after Tomson *et al.* 1981)

Class	
Chloroalkanes	70
Chloroaromatics	94
Alkylbenzenes	98
Alkylphenols	85
Alkyl-naphthalenes	100
Alkanes	71
Alcohols	95
Ketones	98
Indoles, Indenes	96
Amides	74
Alkoxyaromatics	91

Data for the transport of organics to groundwater exist from two slow-rate land treatment sites, a prototype system operated by the U. S. Army Cold Regions Research and Engineering Laboratory in New Hampshire (Jenkins and Palazzo 1981) and the Muskegon system in Michigan. The New Hampshire prototype site showed good removals for natural chloroform and toluene in the wastewater, and greater than 98% removal for four spiked volatile organics (Table 6). At the Muskegon system (Demirjian *et al.* 1983) the irrigation water contained 1-2 $\mu\text{g/l}$ levels of chloroform, 1,2-dichloroethane, 2-chloroaniline, and 2,2'-dichloroazobenzene after lagoon pretreatment. Only two organic compounds were detected as residuals in the soil: xylene, from the dispersing agent used with herbicides, and 2,2-dichloroazobenzene, less than 0.15 mg/kg and possibly derived from photochemical conversion of 2-chloroaniline. There was no evidence for accumulation of

Table 6. Removal of Volatile Organics at a Prototype Slow Rate Land Treatment Site (after Jenkins and Palazzo 1981)

Substance	Mean Concentration ($\mu\text{g/l}$)			
	Wastewater Before Spraying	Wastewater After Spraying	Test Cell 1 Percolates	Test Cell 6 Percolates
Chloroform	41.8	14.0	0.86	0.73
Toluene	57.3	24.4	0.06	0.02
Methylene chloride	7.61	2.32	0.06	0.04
1,1-Dichloroethane	30.2	9.88	b.d.*	0.06
Bromodichloromethane	11.1	3.98	b.d.*	0.02
Tetrachloroethylene	61.9	22.7	0.08	0.35

*Below a detection limit of about $0.01 \mu\text{g/l}$.

other organics after seven years of operation. In the drain tile effluent (at 5-12 feet depth) there was sporadic occurrence of approximately $1 \mu\text{g/l}$ levels of chloroform, 1,2-dichloroethane, and 2-chloroaniline, the latter probably from lagoon seepage. Also in the effluent were $1 \mu\text{g/l}$ levels of the herbicides simazine and atrazine.

From these data it is evident that toxic organics can be transported to groundwater below land treatment sites, although the degree can be controlled by the level of preapplication treatment and application rate as well as choice of effluent and site characteristics. This certainly is cause for concern, but it should be kept in mind that groundwater is not the pristine substance it was once thought to be (Burmaster 1982). The synthetic organic compounds most commonly found in groundwater in the U. S., deriving primarily from industrial wastes, are (Environmental Health Letter 21(6):7, 1982):

trichloroethylene	benzene
tetrachloroethylene	chlorobenzene
carbon tetrachloride	dichlorobenzene
1,1,1-trichloroethane	trichlorobenzene
1,2-dichloroethane	1,1-dichloroethylene
vinyl chloride	cis-1,2-dichloroethylene
methylene chloride	trans-1,2-dichloroethylene

Once organic compounds gain access to groundwater, they may be subject to some of the same removal processes that affect them at the surface, particularly adsorption and microbial decomposition, although certainly at much lower rates. These two processes, which largely govern the movement and fate of organics in the subsurface environment, have been reviewed by McCarty *et al.* (1980, 1981). The degree of adsorption of an organic compound in groundwater is to a great extent dependent upon its hydrophobicity, especially when the aquifer organic content is above about 0.1%. Thus, only compounds with octanol/water partition coefficients less than 10^3 are likely to readily move through the subsurface environment. Of course, these are the very compounds most likely to reach the groundwater at a land treatment site, the more hydrophobic compounds having been adsorbed to the soil above. Likewise, it is probable that most microbial decomposition would have occurred before the organics reach the groundwater, although there is evidence that diverse microbial populations of sulfate reducers, methanogens, and heterotrophs exist and are metabolically active in aquifers, and that biodegradation of some organic pollutants occurs in groundwater (Gerba and McNabb 1981). Nevertheless, it is difficult to avoid the conclusion that once toxic organics get into the groundwater they will remain there for a long time.

Plants

At the low concentrations found in the soil at municipal wastewater land treatment sites, very few organic compounds are likely to be toxic to plants. In a review of data on over 130,000 chemicals, Kenaga (1981) found only 0.17% of the chemicals killed seeds or seedlings at concentrations of 0.1-0.99 ppm. Crop plants, however, although not injured themselves, may accumulate organics that may be toxic to the animals to which they are fed or to humans who use them as food, either directly or through animal products.

Among the organics, the pesticides appear to be the most notorious accumulators in crop plants. Thus, heptachlor, dieldrin, and chlordane are absorbed at low levels from the soil (Braude *et al.* 1978). Most herbicides, of course, are readily taken up and translocated within plants, but there is no reason to think that herbicides would present more of a problem at land treatment sites than they do at ordinary agricultural sites.

In contrast with pesticides, most organic compounds are only poorly absorbed and translocated by plants, with much of the "absorption" probably accounted for by root adsorption. The literature, however, in this area is sparse. Irrigation of vegetables in test plots with contaminated wastewaters has shown no accumulation of polycyclic aromatic hydrocarbons, especially benzo(a)pyrene (Il'nitskii *et al.* 1974). Trace levels of polychlorinated biphenyls (PCBs) from municipal sludge applied to an old field has resulted in no detectable PCBs in plant samples (Davis *et al.* 1981). Higher levels (50-100 ppm dry soil) resulted in 3-50% of the soil concentration in carrots (Iwata *et al.* 1974), with concentration increasing with lesser chlorinated biphenyls. Since 97% of the PCB was found in the carrot peel, very little translocation occurred in the plant tissue. On the basis of greenhouse and field studies of polybrominated biphenyls (PBBs), it has been concluded that little, if any, PBB will be translocated from contaminated soil to plant tops, and although some root crops from highly contaminated soil might contain traces of PBB, much of this PBB could probably be removed by peeling (Chou *et al.* 1978). 4-Chloroaniline and 3,4-dichloroaniline can be absorbed by tomato plants, oats, barley, and wheat, but 90-95% remains in the roots (Fuchsbichler *et al.* 1978); in carrots, however, the chloroanilines are translocated to the upper parts of the plants in significant quantities. In a study of aldehydes and other organics at agricultural land treatment sites Dodelina *et al.* (1976) found no uptake of acetaldehyde, crotonaldehyde, and benzaldehyde in the aboveground portions of potatoes and corn. Cyclohexanone and cyclohexanol could be found in corn plants four days after irrigation, but not later. Dichloroethane was taken up by beets and cereals, but was metabolized and absent within about two weeks after irrigation.

At the operating land treatment site in Muskegon, corn crop samples for 1980 did not contain detectable levels of any of the chemicals tested, and it was concluded that plant uptake of irrigated organic chemicals does not occur to any measurable extent (Demirjian *et al.* 1983).

Animals

The low levels of toxic organics to be expected in the aboveground portions of plants growing at land treatment sites probably pose little hazard to animals feeding upon them. Under certain site-specific conditions, however, high concentrations of particular organics in the wastewater may cause problems. For example, PCBs in cabbage grown on sludge-amended soil have probably caused degenerative changes in liver and thyroid of sheep (Kienholz 1980), and one can extrapolate a similar phenomenon to a land treatment site.

A more serious route of exposure by animals to toxic organics is the soil itself. Most grazing animals ingest a certain amount of soil together with their food plants. Thus, dairy cows may ingest 100-500 kg of soil per year, with an average of about

200-300 kg/yr; expressed in other terms, dairy cows may consume soil up to 14% of dry matter intake when available forage is low and no supplemental feed is used (Kienholz 1980, Fries 1980). These conditions, however, would probably not be characteristic of a land treatment site. Lipophilic organics present in the soil may concentrate in animal fat. For example, feeding experiments with PCBs indicate that the steady-state milk fat concentrations are about five times the diet concentrations, which could result in milk fat levels of 0.7 ppm for each 1 ppm of PCBs in surface soil (Fries 1980). Body fat levels would be expected to be similar.

Conclusions and Research Needs

The tremendous number of organic compounds possibly present in wastewater, together with their myriad health effects and poorly understood behavior in the environment, represent a considerable potential for adverse health effects. Most of these can probably be prevented by simple design and monitoring measures; this, of course, would not be true in the case of high discharges of particular chemicals.

Preapplication treatment by storage lagoons may remove considerable quantities of organics, but cannot be relied upon to efficiently remove all toxic organics, particularly since most pretreatment design questions center on inactivation of pathogens.

Although removal rates of organics from wastewater by aerosolization and volatilization are high, exposure through this route is unlikely to present any significant health effect.

Toxic organics can enter the groundwater, particularly at rapid infiltration sites, and the application and soil factors controlling this transport, together with the factors governing their movement and decomposition within groundwater, are significant research needs.

The levels of toxic organics likely to be present in soils at land treatment sites will probably result in extremely low levels in above-ground portions of plants, but levels in roots, tubers, and bulbs may present a health hazard. The feeding of land-treatment-site-grown plants to animals is unlikely to pose a health problem, but grazing animals may accumulate significant levels of toxic organics. The issue of accumulation of organics from the soil by plants and animals (particularly into milk) is poorly understood, and more research is required.

TRACE ELEMENTS

Types and Levels in Wastewater

The trace elements (including the "heavy metals") in wastewater of public health concern, i.e., those for which primary drinking water standards (USEPA 1977) exist (but excluding silver since its effect is largely cosmetic), are:

Primary Drinking Water Standard (mg/l)	
Arsenic (As)	0.05
Barium (Ba)	1.0
Cadmium (Cd)	0.010
Chromium (Cr)	0.05
Lead (Pb)	0.05
Mercury (Hg)	0.002
Selenium (Se)	0.01

Of these, cadmium, lead, and mercury are usually regarded as of most concern, and barium of minor concern. Chromium and selenium are essential elements in man; arsenic and cadmium have been shown to be essential to experimental animals and, thus, may be essential to man as well (National Research Council 1980). Secondary drinking water standards (USEPA 1979), i.e., those related to aesthetic quality, also exist for copper, iron, manganese, and zinc. These latter elements, as well as all other trace elements, are toxic if ingested or inhaled at high levels for long periods (Underwood 1977), but this fact does not warrant considering them in the land treatment context, where low levels are expected.

Arsenic is popularly known as an acute poison, but chronic human exposure to low doses, as might be expected for all trace elements as a result of land treatment, may cause weakness, prostration, muscular aching, skin and mucosal changes, peripheral neuropathy, and linear pigmentations in the fingernails. Chronic arsenic intoxication may result in headache, drowsiness, confusion, and convulsions (Underwood 1977). Epidemiological evidence has implicated arsenic as a carcinogen, but there is little evidence that arsenic compounds are carcinogenic in experimental animals (Sunderman 1977). Even with high concentrations in soil, however, plants rarely take up enough of the element to constitute a risk to human health (Underwood 1977, Council for Agricultural Science and Technology 1976).

Barium has a low degree of toxicity by the oral route. Because of its effect of intensely stimulating smooth, striated, and cardiac muscle in acute exposures, however, it may have cardiovascular effects in low doses, but this has not thus far been demonstrated (Brenniman *et al.* 1979).

Cadmium is widely regarded as the trace element of most concern from a human health effects viewpoint in the land application of sludge, and this status probably carries over into the land treatment of wastewater as well. The critical health effect of chronic environmental exposure via ingestion is renal tubular damage due to accumulation of cadmium in the kidney. The initial consequence of this damage is the loss of low molecular weight serum proteins in the urine, followed by loss of other proteins, glucose, amino acids, and phosphate. This kidney damage is often irreversible and constitutes a significant adverse health effect (Ryan *et al.* 1982). There is evidence that the absorption and/or toxicity of cadmium are antagonized by

zinc, selenium, iron, and calcium (Sandstead 1977). The carcinogenicity of cadmium is controversial; the epidemiological evidence is tenuous, and the experimental evidence is conflicting (Ryan *et al* 1982).

Chromium is much more toxic in its hexavalent form than its trivalent form, its predominant state in wastewater and soil. Chronic oral exposure in experimental animals has been associated with growth depression, and liver and kidney damage (Underwood 1977). Hexavalent chromium causes respiratory cancer upon chronic exposure to chromate dust (Sunderman 1977). Most crops absorb relatively little chromium from the soil (Council for Agricultural Science and Technology 1976).

Lead chronic toxicity is characterized by neurological defects, renal tubular dysfunction, and anemia. Damage to the central nervous system is common, especially in children, who have low lead tolerance, resulting in physical brain damage, behavioral problems, intellectual impairment, and hyperactivity. At soil pH above 5.5 and high labile phosphorus content, common conditions at a land treatment site, little movement of lead from the soil into plant tops and seed would be expected (Council for Agricultural Science and Technology 1976, Stewart 1979).

Mercury in low levels can result in neurological symptoms such as tremors, vertigo, irritability, and depression, as well as salivation, stomatitis, and diarrhea. Mercury can enter plants through the roots, and appears to be readily translocated throughout the plant (Council for Agricultural Science and Technology 1976), although there is some contrary evidence (Stewart 1979).

Selenium exposure in its chronic form is associated with dental caries, jaundice, skin irritations, chronic arthritis, diseased finger and toenails, and subcutaneous edema. It has also been found to have an inhibitory effect against several types of cancer (Fishbein 1977). Selenium is readily taken up by plants and passed on to animals, and has caused toxicity in livestock in high-selenium soils (Council for Agricultural Science and Technology 1976, Underwood 1977).

Ranges of levels of trace elements in untreated municipal wastewater in the United States are presented in Table 7, together with recommended irrigation water quality criteria for comparison. It is evident that the trace elements in wastewater most likely to violate agricultural irrigation criteria are cadmium and chromium. The high levels of these two elements found in certain municipal wastewaters are doubtlessly due to industrial sources, for example electroplating operations.

Since chromium is poorly absorbed from the soil by crops, cadmium is probably the element of most public health concern. That the upper limits of the cadmium ranges appearing in Table 7, i.e., 0.14 and 1.80 mg/l, might be unusual for municipal wastewaters is suggested by recent data for Chicago (Lue-Hing *et al* 1980), a highly industrialized city. In 1977 five wastewater treatment plants in the Metropolitan Sanitary District of Greater Chicago had average raw wastewater cadmium concentrations of 0.045, 0.021, 0.005, 0.011, and 0.018 mg/l, all below the less restrictive 20-year irrigation criterion of 0.050 mg/l. Purely domestic wastewater, i.e., with no industrial input, in the Chicago area has cadmium concentrations of 0.0011-0.0022 mg/l, of which 0.0002-0.0013 mg/l is due to the local tap water (Gurnham *et al* 1979).

Preapplication Treatment

The removal of trace elements from wastewater by primary treatment (screen, grit chamber, and sedimentation tank) has been reported (Crites and Uiga 1979) to be approximately:

Cadmium	30%
Chromium	40%
Lead	50%

One would expect wastewater stabilization ponds to achieve at least as high removal rates.

Table 7. Concentration of Trace Elements in Untreated Municipal Waste water in U.S. and Recommended Irrigation Water Criteria (mg/l)

Element	Untreated Wastewater		Irrigation Criteria ¹	
	USEPA 1981	Feiler 1980	All Soils ²	Fine-Textured Soils ³
Arsenic	0.003	0.002-0.080	0.10	2.0
Barium	—	—	—	—
Cadmium	0.004-0.14	0.001-1.800	0.010	0.050
Chromium	0.02-0.700	0.008-2.380	0.10	1.0
Lead	0.05-1.27	0.016-0.935	5.0	10.0
Mercury	0.002-0.044	0.0002-0.0039	—	—
Selenium	—	0.001-0.020	0.020	0.020

¹NAS-NAE 1972.

²For waters used continuously on all soils.

³For use up to 20 years on fine-textured soils of pH 6.0 to 8.5.

Soil and Plants

Most of the trace element content of wastewaters appears to be associated with finely-divided suspended solids (Brown 1978, Chang and Page 1978). These are removed near the soil surface by straining and filtration. The removal of the soluble portion depends upon the texture, clay, and organic matter content of the soil, as well as the chemical properties of the specific element, with most soils possessing the capacity to immobilize trace elements near the surface by precipitation reactions and adsorption. Cation exchange reactions do occur, but are not expected to play a major role because of the concentration effect in competition for exchange sites. In any case, cation exchange results in temporary removal from the soil solution. Precipitation reactions include the formation of poorly-soluble oxides, hydroxides, carbonates, phosphates, sulfites, etc., for the cations, and formation of anions for arsenic and selenium. Mercury, of course, may leave the soil through volatilization. The soil chemistry of most of the individual toxic trace elements has been concisely summarized by Chang and Page (1978).

Limits for the maximum cumulative application of trace elements to agricultural land have been recommended by various governmental agencies, for the protection of public health and the prevention of phytotoxicity. These have almost invariably been proposed in the context of the land application of sludge, but should be just as valid for land treatment of wastewater, at least in the slow rate mode. Limits for rapid infiltration and overland flow could be less restrictive because of the lack of production of crops for human consumption and the greater depth of soil involved in treatment (in the former case). These limits have been used, together with typical wastewater levels of trace elements, by Page and Chang (1981) to predict the useful life of a typical land treatment site where crops are grown for human consumption. The results appear in Table 8. It is evident that cadmium, with a 17-67 year limit, is the element most likely to restrict the use of wastewater for irrigation of crops for human consumption. In the case of crops not for human consumption, other elements may be limiting—in particular molybdenum because of its toxicity to livestock, and nickel because of its phytotoxicity. These latter elements yield limits of 47-48 years in Page and Chang's analysis.

Trace elements, of course, are conservative materials, in contrast to organics and pathogens, which may become inactivated and decomposed. Thus, one would expect cadmium to build up in the soil at a land treatment or irrigation site. Our major hope, from a health effects point of view, is that it will become immobilized in the soil, and less available for plant uptake as time passes. There is some suggestion of this in Hinesly's data (Hinesly *et al* 1979) on cadmium content of corn grain and leaves, grown during and after the termination of wastewater sludge application (Table 9). It is of interest to note that the cadmium content of the grain approached that of control

Table 8. Annual Input of Trace Elements and Years of Land Treatment Required to Exceed Recommended Cumulative Input Limits (modified from Page and Chang 1981)

Element	Concentration in Wastewater (mg/l)	Annual Input ¹ (kg/ha)	Recommended Cumulative Input Limits (kg/ha)		Years Required to Exceed Limits	
			USA ²	UK ³	USA	UK
Arsenic	0.005	0.075	—	10	—	133
Cadmium	0.02	0.3	5/10/20 ⁴	5	17/33/67 ⁴	17
Chromium	0.05	0.75	—	1000	—	1333
Lead	0.2 ⁵	3.0	800	1000	267	333
Mercury	0.0009	0.014	—	2	—	143
Selenium	0.005	0.075	—	5	—	67

¹Assuming an annual application rate of 1.5 m.

²USEPA, USFDA, and USDA 1981.

³National Water Council 1977.

⁴For soils with cation exchange capacities of <5, 5-15, and >15 meq/100g, respectively, and soil pH >6.5. If soil pH <6.5, first figure holds.

⁵Raised over the value presented in Page and Chang (1981), to reflect the data in Table 7.

grain within three years after termination of sludge applications, even though the cumulative cadmium application was 58.3 kg/ha, almost three times the maximum recommended limit of 20 kg/ha. The issue of cadmium uptake by plants after annual vs. cumulative application limits and the effect of discontinuing application remain unsettled, however, (Ryan *et al.* 1982).

Examination of trace elements in soil and plants at long-term operating municipal land treatment sites has yielded mixed results. Three locations in the U. S. where crops (in two sites) have been irrigated for 17-33 years showed little or no accumulation of trace elements in soil: Roswell, New Mexico, slow rate site (Koerner and Haws 1979a), Tooele, Utah, slow rate site (Reynolds *et al.* 1979), and Vineland, New Jersey, rapid infiltration site (Koerner and Haws 1979b). After 18 years of slow-rate irrigation with wastewater at San Angelo, Texas, there was no significant change in total soil cadmium concentration, but a six-fold increase of total soil lead concentration (Hassner *et al.* 1978). Wastewater concentrations were <0.004-0.017 mg/l cadmium and <0.050-0.230 mg/l lead, at the low end of the ranges given in

Table 9. Cadmium Uptake by Corn During and After Wastewater Sludge Application (after Hinesly *et al.* 1979)

Year	Sludge Applied (t/ha)	Cd (kg/ha)	Cd in Grain (μg/g)		Cd in Leaves (μg/g)	
			Control	Sludged	Control	Sludged
1969	16.4	7.9	—	—	—	—
1970	52.8	22.6	—	—	—	—
1971	57.8	13.2	0.15	1.37	0.5	35.6
1972	44.4	7.8	0.15	0.89	0.5	23.3
1973	61.1	6.8	0.16	0.44	0.3	7.1
1974	0	0	0.18	0.23	0.1	3.6
1975	0	0	0.15	0.17	0.7	5.9
1976	0	0	0.14	0.15	0.3	2.9
1977	0	0	0.10	0.07	0.3	2.1

Table 7. Slightly lower levels of cadmium in wastewater (<0.001 - 0.008 mg/l) led to a doubling of soil cadmium concentration after 30 years of rapid infiltration at Hollister, California (Pound *et al.* 1978). That trace elements can increase in plants at land treatment sites is documented by results at Werribee Farm in Melbourne, Australia, where, after over 70 years of primary effluent irrigation trace element levels in both soil and plants have increased with cumulative input loads (Croxford 1978). Wastewater, soil, and pasture plant levels of toxic trace elements are summarized in Table 10. Similar levels of cadmium, but lower levels of chromium, have been found in vegetables grown at a site in Santiago, Chile, where the city's untreated wastewater has been applied for over 40 years (Schalscha *et al.* 1978).

Groundwater

Slow-rate land treatment appears to stabilize trace elements, and prevent their entry into the groundwater. Thus, after a year of application of cadmium, copper, lead, nickel, and zinc-amended wastewater to soil columns in the laboratory, Brown *et al.* (1983) found almost all of the metals to remain within the top 25 cm, and none to pass into the leachate at 1.5 m depth. At the prototype land treatment system in Hanover, New Hampshire, trace elements were found to be removed quickly in the first several centimeters of soil, and did not seem to move deeper into the profile, even after five years following termination of the trace element spiking into the wastewater (Jenkins and Palazzo 1981). After 18 years of slow-rate irrigation with wastewater at the San Angelo site, concentrations of cadmium, chromium, and lead were below drinking water standards in seepage creeks, shallow ground wells, and deep wells within the sewage farm (Hossner *et al.* 1978).

Rapid-infiltration sites, in contrast, appear to present some threat to groundwater. After 30 years of primary wastewater infiltration at the Hollister site, concentrations in shallow groundwater of cadmium (0.028 mg/l) and lead (0.09 mg/l) were above drinking water standards, but arsenic, barium, chromium, mercury, and selenium were below (Pound *et al.* 1978).

Animals

The accumulation of trace elements in cattle grazed on sludge-amended pastures has revealed raised levels in liver and kidney, but not in muscle tissue (Bertrand *et al.* 1981a). No increases were seen when cattle were fed sludge-amended-soil-grown forage sorghum (Bertrand *et al.* 1981b). It may not be fair, however, to extrapolate from sludge amended land, where high trace element soil levels may be expected, to land treatment sites. Experience at Werribee Farm in Melbourne, Australia, where cattle are grazed on wastewater-irrigated pastures, has shown higher organ levels of cadmium and chromium than in Farm cattle grazed on non-irrigated pastures, but comparable to non-Farm cattle (Table 11). Organ levels of lead, however, did not increase, in spite of increases in both soil and pasture plants (see Table 10).

Since trace elements accumulate in very small quantities in animal muscle tissue, there is probably little concern about non-visceral meats in the marketplace. Liver and kidneys of animals do, however, accumulate high levels of cadmium, just as they do in man, so that these meats may be of concern to those people consuming large quantities of them.

Conclusions and Research Needs

It seems reasonable to conclude that cadmium is the only trace element likely to be of health concern to humans as a result of land treatment of wastewater, with the exposure being through food plants or organ meats. Groundwater is unlikely to represent a threat except at rapid infiltration sites.

Table 10. Wastewater, Soil, and Pasture Plant Levels of Toxic Trace Elements at Werribee Farm (after Croxford 1978)

Trace Element	Raw Wastewater (mg/l)	Soil ($\mu\text{g/g}$ dry wt)			Pasture Plants ($\mu\text{g/g}$ dry matter)		
		Normal (mean & range)	Farm Control (0-5 cm)	Farm Irrigated (0-5 cm)	Normal	Farm Control	Farm Irrigated
Cadmium	0.015	0.06 (0.01-7)	0.5	0.5-3.9	0.2-0.8	0.19	1.1
Chromium	0.04	100 (5-3000)	13-22	90-325	0.2-1.0	2.3	15
Lead	0.03	10 (2-200)	11-29	56-241	0.1-10	3.4	12

Table 11. Toxic Trace Element Concentrations in Cattle Liver and Kidney at Werribee Farm ($\mu\text{g/g}$ dried tissue) (after Croxford 1978)

	Cadmium	Chromium	Lead
<i>Cattle Liver</i>			
Non-Farm	0.76	0.05	0.5
Farm: Non-Irrigated	0.17	0.05	0.93
Farm: Irrigated	0.38	0.07	1.12
<i>Cattle Kidney</i>			
Non-Farm	3.32	0.06	0.32
Farm: Non-Irrigated	1.24	0.05	2.24
Farm: Irrigated	2.07	0.07	1.41

The significance of this concern with cadmium getting into the human food chain depends upon the cadmium levels presently existing in human food, the total dietary intake of cadmium, and the potential increase in cadmium levels in human food due to land treatment.

The cadmium levels presently existing in human food can be estimated, at least for the United States, by data from the U. S. Food and Drug Administration's Compliance Program ("market-basket survey"). These levels, together with the calculated normal dietary intake and vegetarian dietary intake of cadmium, are summarized in Table 12. It should be noted that root and leafy vegetables have the highest concentrations of cadmium. More accurate estimates of cadmium (and other trace element) concentrations in crops grown in the U. S., together with concentrations in the soils in which they are growing, will soon be available from a survey jointly supported by the USEPA, USFDA, and U. S. Department of Agriculture. In this survey 6,000 crop samples and 18,000 soil samples are being analyzed over a four year period, and the results should be available in the near future.

The present total dietary intake of cadmium was estimated (Table 12) to be about 28 μg /day. Other estimates based on the market-basket method have resulted in higher values: 26-61 μg /day in 15-20-year old U.S. males, by the USFDA, and 52 μg /day in Canadians (Kirkpatrick and Coffin 1977).

A more direct, and potentially more accurate, method of estimating dietary intake of cadmium is by measuring the cadmium content of human feces. This method is feasible because the absorption of cadmium from the gut is low—rarely more than 10%, and usually 4-6%—and the excretion of cadmium into the gut is also very low. It is more accurate because cadmium concentration is generally about ten times more concentrated in feces than food, and because feces reflects actual food intake rather than predicted. A recent study, using existing fecal cadmium data in Chicago and Dallas, and estimating daily feces production, resulted in a final estimate of the average daily intake of cadmium in food for U. S. inhabitants of 13-16 μg /day (Kowal *et al.* 1979). (Since the ingestion rate of the teenage male is often used in discussions of cadmium intake, values of 24 μg /day, 19 μg /day, and 18 μg /day for 10-19-year-old males from Chicago 1974, Chicago 1976, and Dallas, respectively, were estimated.)

This estimate of the average daily intake of cadmium in food can be compared with other estimates by the fecal analysis method, where the daily feces production of each individual was measured rather than estimated. In Sweden rates of 16 μg /day in nonsmokers and 19 μg /day in smokers (former and present) have been reported (see Kowal *et al.* 1979 for references). The increased rate for smokers was partly attributed to their increased food intake. Nine μg /day fecal cadmium has been

Table 12. Cadmium Concentration in Foods and Calculated Dietary Intake (from Ryan *et al.* 1982)

Food Classes	ppb Cd ^a	Normal Diet ^b		Vegetarian Diet ^c	
		g/day	µg Cd/day	g/day	µg Cd/day
Dairy products	5.7	549	3.1	584	3.3
Meat, fish, poultry	15.3	204	3.1	—	—
Grain & cereal products	23.2	331	7.7	203	4.7
Potatoes	48.0	138	6.6	43	2.1
Leafy vegetables	40.5	42	1.7	252	10.2
Legume vegetables	6.2	51	0.3	166	1.0
Root vegetables	32.3	25	0.8	—	—
Garden fruits	14.7	69	1.0	—	—
Fruits	3.0	173	0.5	284	0.8
Oily fats, shortenings	15.3	56	0.9	107	1.6
Sugars & adjuncts	10.0	65	0.7	110	1.1
Beverages	3.0	534	1.6	600	1.8
Total Intake		2,237	28.0	2,349	26.6

^aFrom FDA Compliance Program Evaluation 1974 Total Diet Studies.

^bAdjusted on a caloric basis from the FDA 1974 Total Diet Studies to represent the normal diet which compares with the adult lacto-ovo-vegetarian diet.

^cLoma Linda lacto-ovo-vegetarian diet. Based on response of 183 southern Californians in a food frequency questionnaire by the Department of Biostatistics and Epidemiology, Loma Linda University School of Health, 1978. Leafy vegetables class includes root vegetable and garden fruit classes from normal diet.

measured in Sweden, compared with a value of 10 µg/day measured by the total diet collection method. In Germany 31 µg/day has been measured, compared with 48 µg/day measured by the market-basket method. In Japan, where cadmium levels in food are higher, the fecal analysis method has resulted in several estimates ranging from 24 µg/day to 84 µg/day.

"It has generally been concluded that ingestion of 200 to 350 mg Cd/day over a 50-year exposure period is a reasonable estimate for individuals (excluding smokers and occupationally exposed) within the population to reach the critical renal concentration (200 mg Cd/g wet weight in the renal cortex) associated with the initiation of proteinuria. This ingestion limit assumes background exposure levels of air and no exposure from smoking. If these exposures are increased, then the suggested ingestion limit must be correspondingly reduced. Smoking one pack of cigarettes/day will reduce the limit by about 25 µg/day. Again these exposures are assumed to occur over a 50-year exposure period and, in the case of cigarettes, since many smokers start as teenagers, this addition would be relevant for much (30 to 35 years) of the 50-year exposure period. Therefore, smokers must be considered as being at increased risk." (Ryan *et al.* 1982).

Thus, present levels of total dietary intake of cadmium for most people appear to be fairly safe. However, in view of human variability in sensitivity and the variability in food supply, these levels probably should not be allowed to rise greatly.

It is of interest to note that increased consumption by individuals of those leafy and root vegetable crops highest in cadmium, and of organ meats as well, would increase the dietary iron intake. Since iron-sufficient humans absorb only about 2.3% of dietary cadmium, compared to an average absorption of 4.8% (in the generally iron-deficient American population), the increased iron intake would tend to correct for the increased cadmium intake (Chang 1980).

The potential increase in cadmium levels in human food due to land treatment or irrigation is still an unsettled question. Almost all the relevant research on the subject has been done with the land application of wastewater sludge, but, in spite of the high cadmium-application rates associated with this practice, insufficient time has elapsed to allow many firm conclusions to be drawn. It is clear, however, that increased cadmium in the soil results in increased cadmium in the plants grown in that soil, the degree of increase being a function of cadmium amendment, plant species and cultivar, soil pH, organic matter, and time since application (Ryan *et al.* 1982). The degree of risk to man, of course, is dependent upon the amount of the food supply affected and the diet selection of the individual.

The most significant research need in the areas of trace elements probably continues to be the development of an understanding of the factors controlling the uptake of trace elements by plant crops at land treatment sites, and their entry into the human food supply.

NITRATES

Nitrogenous wastes are important constituents of municipal wastewaters, consisting of (1) proteins and other nitrogenous organics from feces, food wastes, etc., (2) urea from urine, and (3) their breakdown products. Raw domestic wastewater has concentrations of about 8-35 mg/l organic nitrogen, 12-50 mg/l ammonium (plus ammonia), and, thus, 20-85 mg/l total nitrogen, all expressed as N (Metcalf and Eddy 1972). Nitrites and nitrates are normally present only in trace amounts in fresh wastewater. Bacteria rapidly decompose most forms of organic nitrogen to ammonium (or ammonia), in wastewater or soil. Under aerobic conditions ammonium is oxidized by bacteria (*Nitrosomonas*) to nitrite, and the nitrite rapidly oxidized by bacteria (*Nitrobacter*) to nitrate; the two-step process is called "nitrification." Under anaerobic conditions, and in the presence of organic matter, bacteria can use nitrate as a source of oxygen, and convert nitrate to molecular nitrogen, which escapes to the atmosphere; this is called "denitrification." Both aquatic and terrestrial plants can use ammonium and nitrate as a nitrogen source.

Inorganic nitrogen is normally quite innocuous from a human health point of view, although high ammonia levels can present an aesthetic problem. The major health concern is that infants, less than about three months of age and consuming large quantities of high-nitrate drinking water through prepared formula, have a high risk of developing methemoglobinemia. The incompletely developed capacity to secrete gastric acid in the infant allows the gastric pH to rise sufficiently to encourage the growth of bacteria which reduce nitrate to nitrite in the upper gastrointestinal tract. The nitrite is absorbed into the bloodstream, and oxidizes the ferrous iron in hemoglobin to the ferric state, in which form it is incapable of carrying oxygen. Fetal hemoglobin (Hb F), 50-89% of total hemoglobin at birth, is particularly susceptible to this transformation. Methemoglobin is normally present in the erythrocytes of adults, at a concentration of about 1% of total hemoglobin, being formed by numerous agents, but kept to a low level by the methemoglobin reductase enzyme system. This enzyme system is normally not completely developed in young infants. At a methemoglobin concentration of about 5-10% of total hemoglobin the body's oxygen deficit results in clinically-detectable cyanosis. As a result of epidemiological and clinical studies (Shuval and Gruener 1977, Craun *et al.* 1981, Fraser and Chilvers 1981) a primary drinking water standard of 10 mg/l of nitrate-nitrogen (i.e., nitrate expressed as N) has been established (USEPA 1977) to prevent this condition from developing.

Besides methemoglobinemia, there is also some concern about nitrates resulting in the formation of carcinogenic N-nitroso compounds in the gut, but this phenomenon probably involves higher concentrations than the 10 mg/l water standard (Fraser *et al.* 1980, Fraser and Chilvers 1981).

The relevance of land treatment, of course, centers on the possibility of highly soluble nitrates reaching groundwater which may be used as a potable water supply.

Preapplication Treatment

Wastewater treatment, planned or otherwise, results in the rapid breakdown of organic nitrogen to ammonium, and the oxidation of ammonium to nitrate, i.e., nitrification. Under anaerobic conditions, for example at the bottom of a wastewater stabilization pond, some denitrification may occur, resulting in nitrogen loss from the wastewater (USEPA 1983). In a pond some nitrogen may also be taken up by algae (DiGiano and Su 1977), but this is not really lost from the wastewater.

During treatment, organic carbon is constantly lost as carbon dioxide, so that, while the C:N ratio in raw wastewater is about 5:1, the C:N ratio in secondary effluent may be about 1:2. Since denitrifying bacteria require a source of organic carbon to supply their energy needs, the level of organic carbon in wastewater is critical to the removal of the highly mobile nitrate ion by denitrification in the anaerobic layers of the soil. Where glucose is used as the carbon source the C:N ratio required by the denitrification reaction is about 3.2:1. Thus, if groundwater is to be protected from nitrate and wastewater application rates are going to exceed the rate of nitrogen uptake by plants (if any), primary treatment alone, i.e., sedimentation, has been recommended as preapplication treatment (Pound *et al.* 1978).

The influence of the C:N ratio on the degree of denitrification in the soil has been demonstrated in numerous field and laboratory studies. For example, in studies on the application of primary and secondary effluent to soil columns with and without vegetation, Lance *et al.* (1980) observed the following nitrogen removals:

	Vegetated Column	Nonvegetated Column
Primary Effluent:	81.8%	45.6%
Secondary Effluent:	48.1%	28.5%

Increased nitrogen removal occurred with higher C:N ratio (i.e., primary effluent) and the presence of vegetation.

Groundwater

The threat of nitrates to groundwater is less at slow-rate than rapid-infiltration sites because of lower application rates and the presence of plant uptake. (Overland flow systems should not affect groundwater.) Nitrogen concentrations in applied wastewater and groundwater at several operating slow-rate systems are summarized in Table 13; nitrogen is reported as total nitrogen, but in wastewater consists mostly

Table 13. Wastewater and Groundwater Nitrogen at Slow-Rate Land Treatment Sites (mg/l as N)

Site	Total Nitrogen Concentration			Reference
	Applied Wastewater	Affected Groundwater	Control Groundwater	
Dickinson, ND	12.1	3.0	1.1	Benham-Blair <i>et al.</i> 1979a
Hanover, NH	27-28	7.3	—	Jenkins & Palazzo 1981
Lubbock, TX	12-15	50	8.5	Hinesly <i>et al.</i> 1978
Roswell, NM	36	5-7	3	Koerner & Haws 1979a
San Angelo, TX	29.8	15.7	22.0	Hossner <i>et al.</i> 1978
Braunschweig, Germany	49.2	32.8	11.6	Tietjen <i>et al.</i> 1978
Wroclaw, Poland	42-45	6-31	—	Cebula & Kutera 1978
Melbourne, Australia	51.3	6.6	—	McPherson 1978
Santiago, Chile	35-38	12-15	2.7	Schalscha <i>et al.</i> 1979

of organic N and ammonium, and in groundwater mostly of nitrate. Of the U. S. sites, only that at Lubbock appears to be overloaded, resulting in a groundwater concentration of 50 mg/l N.

Nitrogen concentrations at several operating rapid-infiltration systems are summarized in Table 14. It is evident that many rapid infiltration systems result in groundwater nitrate concentrations above the 10 mg/l drinking water standard. Many of these systems could improve their performance by decreasing the loading rate (thus, allowing more time for within-soil treatment), increasing the C:N ratio (particularly by using primary effluent instead of secondary), and/or optimizing the flooding-drying sequence. Leach and Enfield (1983), for example, have found an operating sequence of one-day flooding and one-day drying to be the optimum regime for total nitrogen removal in their experimental system. If it is not feasible to prevent the groundwater beneath a rapid-infiltration site from rising above the nitrate standard, it may be possible to ensure that the water is not used as a drinking water supply. Rapid infiltration basins could be located where groundwater flows into surface waters, or the percolate could be recovered with wells or underdrains for surface discharge or reuse in crop irrigation (Reed 1979).

It should be kept in mind that land treatment sites are not the only source of nitrate in groundwater. Many groundwaters are naturally high in nitrates, e.g., that in the vicinity of San Angelo, Texas (Table 13), and in urban areas on-site absorption fields and lawn fertilizers have been shown to be sources of nitrates in groundwater (Porter 1980).

Conclusions and Research Needs

Land treatment systems, particularly rapid-infiltration, threaten to raise the nitrate concentration in their underlying groundwater above the drinking water standard of 10 mg/l as N. This can be prevented, however, by proper siting and management practice, e.g., using high C:N ratio wastewater, matching loading rate to crop uptake (for slow-rate systems), and optimizing the flooding-drying regime. These management practices and the agronomic factors controlling the entrance of nitrates into groundwater are important research needs.

Table 14. Wastewater and Groundwater Nitrogen at Rapid-Infiltration Land Treatment Sites (mg/l as N)

Site	Loading Rate (m/yr)	Total Nitrogen Concentration			Reference
		Applied Wastewater	Affected Groundwater	Control Groundwater	
Boulder, CO	48.8	16.5	9-16	—	Smith <i>et al.</i> 1979
Brookings, SD	12.2	10.9	6.2	—	USEPA 1981
Calumet, MI	17.1	24.4	7.1	—	USEPA 1981
Fort Devens, MA	30.5	50	19.6	—	USEPA 1981
Hollister, CA	15.2	40.2	1.7-7.8	0-9.9	Pound <i>et al.</i> 1978
Lake George, NY	58	12	8	—	Aulienbach 1979
Milton, WI	244	26.3	15.2	6.9	Benham-Blair <i>et al.</i> 1979b
Phoenix, AZ	61	27.4	9.6	—	Bouwer <i>et al.</i> 1980
Vineland, NJ	—	34-41	17-29	2.9	Koerner & Haws 1979b

SODIUM

Sodium may enter the groundwater beneath land treatment sites, just as nitrogen does. For example, at the the Wroclaw, Poland, sewage farm, an untreated wastewater sodium content of 83-115 mg/l was reflected in a groundwater content of 90-144 mg/l (Cebula and Kuter 1978). This level may be compared with the sodium content of U. S. drinking waters, 58% of which have 0-20 mg/l, and only 14% of which have over 100 mg/l (White *et al.* 1967). A primary drinking water standard for sodium has not been established (USEPA 1977).

The health significance of these raised levels in groundwater, and thus potentially in drinking water, is unclear, particularly since drinking water sodium is a small portion of total dietary intake (2-8 g/day). That a decreased intake of sodium can reduce blood pressure in subjects with mild hypertension has been shown by several clinical studies, e.g., Beard *et al.* (1982), but the overall significance of restricted sodium consumption to preventing hypertension in the general public has been recently challenged (Kolata 1982, Boffey 1982, Puska *et al.* 1983, Rouse *et al.* 1983). Similarly, the evidence that high sodium consumption causes hypertension is tenuous. However, increased groundwater sodium concentrations beneath land treatment sites should be kept in mind as a possible future health concern.

COMPARISON WITH CONVENTIONAL SYSTEMS

The comparison of the potential health effects of land treatment, caused by both pathogens (Kowal 1982) and toxic substances, with those of conventional treatment is necessarily highly subjective. Nevertheless, there are suggestions that land treatment is at least equally protective of public health as conventional treatment.

A comparison of bacterial aerosol levels at conventional activated sludge plants and a spray irrigation land treatment site has been performed by Clark *et al.* (1978), to evaluate relative human exposure levels at these two types of facilities. They concluded that airborne bacterial levels, as measured by fecal coliforms, appear to be higher at the activated sludge plants than at the spray irrigation facility.

A broad comparison of health risks between activated sludge treatment and slow-rate land treatment has been performed by Crites and Uiga (1979, Uiga *et al.* 1978). The comparison assumed: (1) A flow of 3 M gal. day of domestic wastewater. (2) Activated sludge treatment is followed by disinfection and surface water discharge. (3) Land treatment is preceded by aerated lagoon preapplication treatment and storage, and followed by percolate water recovery using underdrains and surface water discharge, with no disinfection. They arrived at the following conclusions comparing the two systems:

1. If maintained and operated properly, both conventional and land treatment systems provide a large measure of safety for public health. Slow-rate land treatment offers greater protection against parasites, viruses, trace organics, halogenated organics, trace elements, and nitrate.
2. Since adequate removal of parasite eggs and cysts require such measures as filtration or long detention times in ponds or storage lagoons, land treatment offers greater protection from health risks.
3. Land treatment systems, especially those with ponds and storage lagoons, remove viruses to a higher degree than do conventional treatment and disinfection systems.
4. Land treatment systems are less susceptible to failure or upsets than conventional systems, especially for small systems.

In an overview of existing land treatment systems, Iskandar (1978) concluded that "the potential health hazards from land treatment are no greater and probably less than those from conventional treatment. . . . Although land treatment, like all known waste treatment systems, has potential health hazards associated with it, these risks can be kept to a minimum." The risks associated with alternative systems of wastewater treatment and disposal, conventional as well as land treatment, should be defined and compared. He considered it to be rather odd that land treatment instills so much more public fear than conventional treatment.

GENERAL CONCLUSIONS

This section contains the general conclusions and research needs from both this report on toxic substances, and an earlier companion report on pathogens, "Health Effects of Land Treatment: Microbiological" (Kowal 1982).

Types and Levels in Wastewater

The types and levels in wastewater of most pathogens are fairly well understood, with the exception of viruses. Since only a small fraction of the total viruses in wastewater and other environmental samples may actually be detected, the development of methods to recover and detect viruses continues to be a research need. The occurrence of viruses in an environmental setting should probably be based on viral tests rather than bacterial indicators since failures in this indicator system have been reported.

The tremendous number of organic compounds possibly present in wastewater, together with their myriad health effects and poorly understood behavior in the environment, represent a considerable potential for adverse health effects. Most of these can probably be prevented by simple design and monitoring measures; this, of course, would not be true in the case of discharges containing high levels of particular chemicals.

It seems reasonable to conclude that cadmium is the only trace element likely to be of health concern to humans as a result of land treatment of wastewater, with the exposure being through food plants or organ meats.

Preapplication Treatment

The level of preapplication treatment required for the protection of public health may be as little as properly-designed sedimentation at land treatment sites with limited public access, where crops are protected by appropriate crop choice and waiting periods, and groundwater is protected by appropriate hydrological studies and application rate selection. Where protection of groundwater cannot be assured, wastewater stabilization ponds should be considered for virus removal, but further investigations into the survival of viruses in these ponds is an important research need, as is that of protozoan cysts. Because of potential contamination of crops and infection of animals, slow-rate and overland-flow systems should have high removal rate of helminth eggs. These relatively simple pretreatment requirements would be appropriate for many land treatment systems in the U. S., e.g., for many slow-rate sites where crops for animal feed are grown.

Preapplication treatment by storage lagoons may remove considerable quantities of organics, but cannot be relied upon to efficiently remove all toxic organics, particularly since most pretreatment design questions center on inactivation of pathogens.

The recommendations made in the paragraphs below assume a minimum level of preapplication treatment, i.e., properly-designed sedimentation. In situations with greater public access (e.g., renovated water reuse on golf courses), shorter waiting periods before grazing or harvest of crops (e.g., agriculture in arid areas), or threat of groundwater contamination (e.g., shallow water table used as a drinking water source), more extensive preapplication treatment may be required. This treatment may consist of wastewater stabilization ponds, conventional treatment unit processes, or even disinfection. The exact degree of pretreatment required for these

situations is site-specific, and recommendations should be determined separately for each system (Lance and Gerba 1978).

Aerosols

Because of the potential exposure to aerosolized viruses at land treatment sites, it would be prudent to limit public access to 100-200 m from a spray source, unless the effluent has been disinfected. At this distance bacteria are also unlikely to pose significant risk. Human exposure to pathogenic protozoa or helminth eggs through aerosols is extremely unlikely.

Suppression of aerosol formation by the use of downward-directed, low-pressure nozzles, ridge-and-furrow irrigation, or drip irrigation is recommended where these application techniques are feasible.

Although removal rates of organics from wastewater by aerosolization and volatilization are high, exposure through this route is unlikely to present any significant health effect.

Surface Soil and Plants

The survival times of pathogens on soil and plants are summarized in Table 15 (after Feachem *et al.* 1978). Since pathogens survive for a much longer time on soil than plants, the recommended waiting periods before harvest are based upon probable contamination with soil.

Aerial crops with little chance for contact with soil should not be harvested for human consumption for at least one month after the last wastewater application; subsurface and low-growing crops for human consumption should not be grown at a land treatment site for at least six months after last application. These waiting periods need not apply to the growth of crops for animal feed, however.

An important research need is the effect of drying of the soil between wastewater applications on the survival of surface-soil viruses.

The levels of toxic organics likely to be present in soils at land treatment sites will probably result in extremely low levels in above-ground portions of plants, but levels in roots, tubers, and bulbs may present a health hazard.

The potential increase in cadmium levels in human food due to land treatment or irrigation is still an unsettled question. It is clear, however, that increased cadmium in the soil results in increased cadmium in the plants grown in that soil, the degree of increase being a function of cadmium amendment, plant species and cultivar, soil pH, organic matter, and time since application (Ryan *et al.* 1982). The degree of risk to man, of course, is dependent upon the amount of the food supply affected and the diet selection of the individual. Present levels of total dietary intake of cadmium for most people appear to be fairly safe. However, in view of human variability in sensitivity and the variability in food supply, these levels probably should not be allowed to rise greatly.

Table 15. Survival Times of Pathogens on Soil and Plants

Pathogen	Soil		Plants	
	Absolute Maximum	Common Maximum	Absolute Maximum	Common Maximum
Bacteria	1 year	2 months	6 months	1 month
Viruses	6 months	3 months	2 months	1 month
Protozoa	10 days	2 days	5 days	2 days
Helminths	7 years	2 years	5 months	1 month

The most significant research need in the area of trace elements probably continues to be the development of an understanding of the factors controlling the uptake of trace elements by plant crops at land treatment sites, and their entry into the human food supply.

Movement in Soil and Groundwater

Properly designed slow-rate land treatment systems pose little threat of bacterial or viral contamination of groundwater. Considerable threat of bacterial contamination exists, however, at rapid-infiltration sites where the water table is shallow, particularly if the soil is porous. The survival of bacteria in groundwater, once they get there, is poorly understood, and is an important research need.

Likewise, considerable potential for viral contamination of groundwater exists at rapid-infiltration sites, and appropriate preapplication treatment or management techniques should be instituted, e.g., intermittent application of wastewater. Until then, groundwater drawn for use as potable water supplies should be disinfected. The factors controlling the migration of viruses in soils, and the survival of viruses in groundwater, are poorly understood, and are significant research needs.

Human exposure to pathogenic protozoa or helminths through groundwater is extremely unlikely.

Toxic organics can enter the groundwater, particularly at rapid infiltration sites, and the application and soil factors controlling this transport, together with the factors governing their movement and decomposition within groundwater, are significant research needs.

Groundwater is unlikely to represent a significant trace element threat except at rapid infiltration sites.

Land treatment systems, particularly rapid-infiltration, threaten to raise the nitrate concentration in their underlying groundwater above the drinking water standard of 10 mg/l as N. This can be prevented, however, by proper siting and management practice, e.g., using high C:N ratio wastewater, matching loading rate to crop uptake (for slow-rate systems), and optimizing the flooding-drying regime. These management practices and the agronomic factors controlling the entrance of nitrates into groundwater are important research needs.

Increased groundwater sodium concentrations beneath land treatment sites should be kept in mind as a possible future health concern.

Animals

There appears to be little danger of bacterial, viral, or protozoan disease to animals grazing at land treatment sites if grazing does not resume until four weeks after last application. However, the role of animals in transmitting human viral diseases at land application sites is poorly known, and is a research need. Removal of helminth eggs during preapplication treatment should eliminate the potential of disease from those long-lived parasites. The feeding of land-treatment-site-grown plants to animals is unlikely to pose a health problem, but grazing animals may accumulate significant levels of toxic organics. The issue of accumulation of organics from the soil by plants and animals (particularly into milk) is poorly understood, and more research is required.

Infective Dose, Risk of Infection, Epidemiology

Because of the possibility of picking up an infection, it would be wise for humans to maintain a minimum amount of contact with an active land treatment site. The comparison of the respiratory infective dose of enteric viruses with the oral infective dose is a significant research need.

Epidemiological studies to date suggest little effect of land treatment on disease incidence. However, well planned and implemented prospective studies have not been completed.

Perhaps most importantly, a comparison of land treatment systems with conventional treatment systems suggests that the former is at least equally protective of public health as the latter.

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